

A novel material design system method for on-demand material development

— A method born from a development field —

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[Translation from *Synthesiology*, Vol.7, No.1, p.36-42 (2014)]

Advanced materials for the semiconductor industry are being developed every month. To reduce the development period, we constructed a novel material design system method based on linear programming, combinatorial optimization, and graph theory. This method (called weak conditioned combinatorial linear programming) enabled us to find formulation composition candidates that satisfy a number of properties at the same time. By defining the solution area as a function of the combination index, the optimum formulations were acquired. This optimization could be done by newly developed user-friendly software. This system is applicable to optimization of materials with complex properties and time-series properties such as creep. The method enabled us to develop materials satisfying target values efficiently.

Keywords : Linear programming, combinatorial optimization, on demand development, material design

1 Introduction

The most important points in material development are long-term steady research and eyes that do not miss any accidental finds (serendipity). While we do not wish to raise objection to this mainstream thinking, it is a fact that new materials must be developed almost every month at the development sites of the state-of-the-art semiconductor materials. There is no time to wait around for the arrival of some novel materials or serendipitous finds, and in reality, there is much struggle and battle to fulfill the target values within a set deadline. With this background, what style of development can the researchers use?

Since such material development is done by combining the materials at hand with the skill of a craftsman, it may fall in the realm of *bricolage* (or tinkering) as described by Lévi-Strauss.^{[1][2]}

However, the authors have engaged in ultra short-term developments for over 10 years, and we started to feel that we wished to develop materials that satisfy the target values through rational design, rather than by mere *bricolage*. This thinking is close to engineering (the way of manufacturing by preparing the necessary tools and making it according to a design plan) that is the counter-concept of *bricolage*. We wished to construct a development method that has both the merits of the creative aspect of *bricolage* and the rational aspect of engineering design.

The style of material development that was generally done and

the style devised by the authors are compared in Fig. 1. The normal material research is carried out to find an innovative material with outstanding property. The research style proposed in this paper is a material design in which the already existing materials are combined and outstanding performance is not sought. For example, in the semiconductor package manufacturing process, while there are diverse target properties such as fluidity and viscosity, it is important for the individual properties to fall in a certain range, as outstanding excellent properties are not sought.

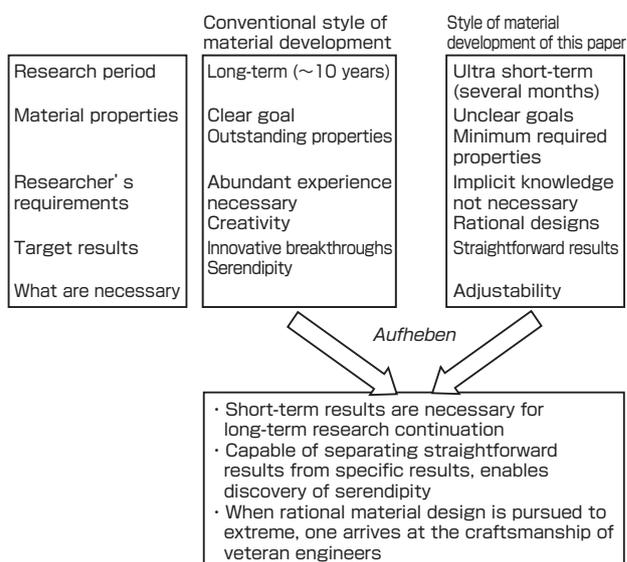


Fig. 1 Comparison of the conventional style of material development and the style described in this paper

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Original manuscript received March 28, 2013, Revisions received September 3, 2013, Accepted September 3, 2013

The greatest characteristic of this style of material development is to accomplish the project in an extremely short time, and therefore, it will be called on-demand material development in this paper. The concept of the on-demand material development method that the authors established and the actual applications will be explained.

2 Background of research

The state-of-the-art electronic devices such as smart phones, tablets, ultra thin laptop PCs, and servers that support the cloud are evolving continuously, and the semiconductor elements and semiconductor packages that are the heart of such devices are also evolving.^[3]

Various materials are used in the semiconductor package as shown in Fig. 2. Since the semiconductor package changes in structure in response to the preference and trend of the consumers, mainly young people (for example, downsizing and increased memory of smart phones), the trend of technological evolution is characteristically difficult to predict. For example, the requirements of the die-bonding film, or the adhesive material for semiconductors used to attach the semiconductor chip to its supporting substrate, change every month.^{[4][5]}

As shown in Fig. 3, the assembly process of the semiconductor package is diverse, and various properties are required for each process. The commonly required properties include:

- (1) It must be fluid at temperature 80 °C or lower, and be able to bond to the wafer.
- (2) Since it is used to bond the chip and the substrate, it must be able to absorb the difference of coefficient of thermal expansion and relieve stress.
- (3) It must not flow or detach during the soldering process (about 260 °C) to attach the semiconductor package to the wiring substrate.

Even a semiconductor manufacturer may not know the required properties of materials at the beginning of the development for a new structure. Rather, several materials are evaluated to

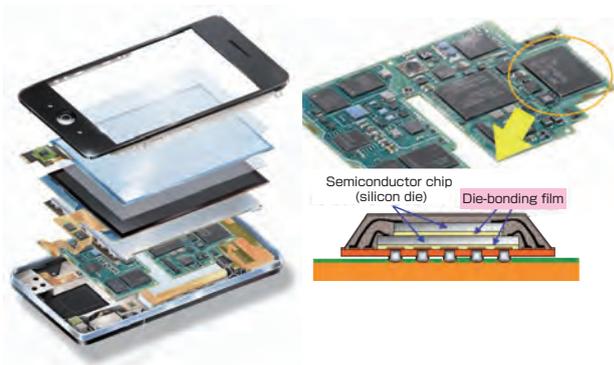


Fig. 2 Examples of an electronic device and a semiconductor package

clarify a target value.

The development period of a material is as short as a few months, but it is extremely difficult to synthesize any new material such as polymers in a few months in a specific form such as films.

On the other hand, it is well known that the state-of-the-art semiconductor industry cannot survive without extremely high facility investments that must be recovered in a short period and then invested for the next generation. In the state-of-the-art semiconductor industry, there is an increased demand for rational design and optimization methods for quick development of materials to strengthen the competitiveness and to reduce the investment risk.

The mathematical optimization methods that have been considered so far include linear programming, nonlinear programming, and combinatorial optimization.^{[6]-[9]} Although these methods are expected to be applied to material design, there are several issues in utilizing them:

- (1) It is necessary to select the optimal combination among several chemical materials, but in practice, the number of materials that can be actually used is limited due to the limitations of the number of tanks for storing and mixing the materials, the number of waste fluid pipes, or the risk of supply cutoff due to natural disasters or plant accidents.
- (2) There are cases where the candidate set of solutions is not a convex set because the constrained function becomes complex due to the limitation of patents held by other companies.
- (3) In designing the adhesive agent, the optimization of the complex physical properties such as the complex elastic modulus is essential. However, the conventional mathematical design targets the real vector space, and the handling of complex numbers has been difficult.
- (4) It is not necessary to find the optimal value for one point

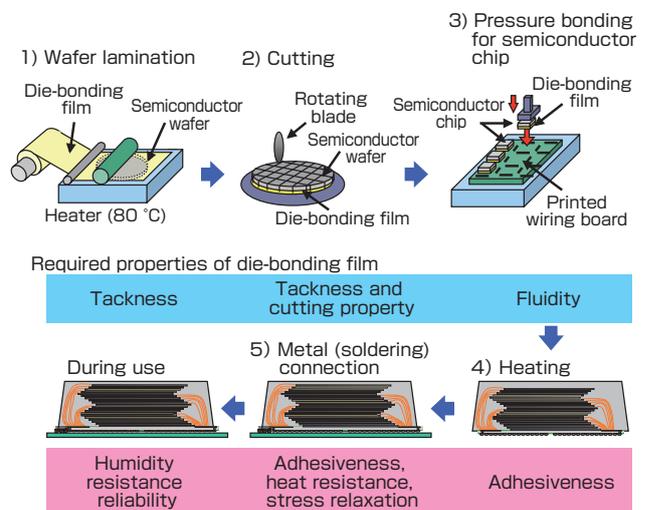


Fig. 3 Assembly process of the semiconductor package

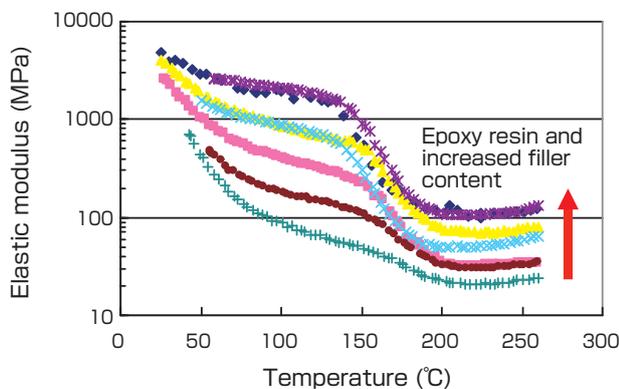
only, but rather, the breadth of solution range that satisfies the target value to prepare for the variations and design change is important.

Therefore, it is necessary to build a mathematical design method to solve these issues and to utilize them in the actual development.

The authors have investigated the design system methods that were appropriate for material design and were readily usable on site. After carefully surveying the correlation between the composition of the materials and their properties, we thought the shortcut was to build a method based on linear programming, because in many cases, a quasi-linear relationship existed between the composition and the material property. However, when this was applied to material development, even if the target function was expressed as the only line, the solution did not necessarily exist. Also, the robustness against the variation of the film property was necessary. This meant that it was important to consider the breadth and form of range that satisfied the target value by weakening the condition of the solution, rather than strengthening the limiting condition of the solution (or consider only the optimal value). Therefore, we developed weak conditioned combinatorial linear programming and software, by applying linear programming, combining multiple materials, and calculating the combination to satisfy the multiple target values.^{[10]-[12]}

3 Basics of the weak conditioned combinatorial linear programming

Die-bonding film is a composite material made by combining various materials including epoxy resin, fillers such as inorganic particles, as well as acryl rubber, and catalysts. By changing the composition ratio, a wide range of changes is possible if focus is placed only on elastic modulus, as shown in Fig. 4a. We are often asked by the semiconductor manufacturers, “The goal is mostly satisfied, so can you just



a) Elastic modulus at various composition

Fig. 4 Correlations among various properties

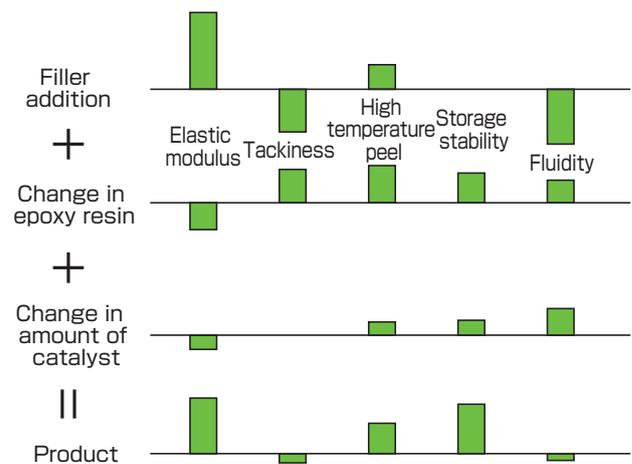
lower the elastic modulus?” However, this is the most difficult request, because when the amount of ingredients is changed, all the property values change, not just the elasticity, as shown in Fig. 4b. By lowering the elasticity, all the properties that had previously satisfied the target values may shift from the acceptable target range.

To solve this issue, the authors constructed a design method based on linear programming as shown in the equation below. The value that is linear mapped by the composition/property matrix from the composition is the property. For example, a_{mn} shows the relationship between the composition content and the adhesiveness of an epoxy resin. The matrix consisting of a_{mn} is the property/composition matrix, one for $(c_1 \dots c_n)$ is the content vector, $(p_1 \dots p_n)$ is the property vector, $(k_1 \dots k_n)$ is the combination vector, and \otimes is the Hadamard product.

$$\begin{pmatrix} p_1 \\ p_2 \\ \vdots \\ p_m \end{pmatrix} = \begin{pmatrix} a_{11} & \dots & a_{1n} \\ a_{12} & & a_{2n} \\ \vdots & \ddots & \vdots \\ \vdots & & \vdots \\ a_{m1} & \dots & a_{mn} \end{pmatrix} \begin{pmatrix} k_1 \\ k_2 \\ \vdots \\ k_n \end{pmatrix} \otimes \begin{pmatrix} c_1 \\ c_2 \\ \vdots \\ c_n \end{pmatrix}$$

The Hadamard product of the matrix is defined as follows.

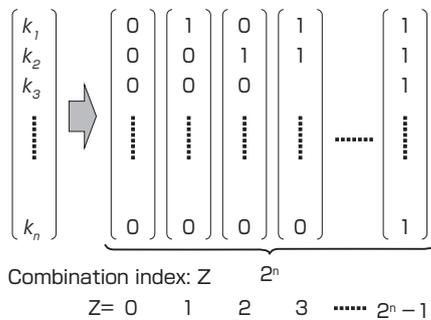
$$A \otimes B = \begin{pmatrix} a_{11} & \dots & a_{1n} \\ \vdots & \ddots & \vdots \\ \vdots & & \vdots \\ a_{m1} & \dots & a_{mn} \end{pmatrix} \otimes \begin{pmatrix} b_{11} & \dots & b_{1n} \\ \vdots & \ddots & \vdots \\ \vdots & & \vdots \\ b_{m1} & \dots & b_{mn} \end{pmatrix} = \begin{pmatrix} a_{11} \cdot b_{11} & \dots & a_{1n} \cdot b_{1n} \\ \vdots & \ddots & \vdots \\ \vdots & & \vdots \\ a_{m1} \cdot b_{m1} & \dots & a_{mn} \cdot b_{mn} \end{pmatrix}$$



b) Conceptual diagram of contribution of various elements to the properties

If the quantities of the materials are changed as shown in Fig. 4b, all properties fluctuate. They can be matched to the target value by offsetting and adding the fluctuations.

The material combination k_n parameter is set as 1 when material n is used, and 0 when it is not used. The material combination vector will be in one-to-one correspondence with the combination index (will be described Z). For example, when considering the composition by selecting 0~ n types of material from n number of materials, there will be 2^n combinations. If the expression of combination is expressed as 010101..., the expression becomes complicated when there are many types of materials. Therefore, the number sequence is set as binary, and one-to-one correspondence is given to decimal Z . The corresponding composition parameter c_n is changed to see whether the property values lie in the target range, and this will enable the check of the composition range that satisfies the target value.



While the conventional linear programming sought the maximum or minimum values of the evaluation function, here, all the solutions that satisfy the target values are considered as the solution set, and therefore, it is called “weak condition.” Weakening the condition of the solution contributes to shortening the computation time.

Mathematically, the weak conditioned combinatorial linear programming creates the space P of the realizable properties, as the space C created by the composition parameter c_n that possesses limitations is linear mapped to the property space by the matrix consisting of the composition/property parameter, as shown in Fig. 5. The product set $S(=P \cap T)$ of space P and space T of target value comprise the space that satisfies the target value. Also, space C_s created by compositional parameter c_n that satisfies S is defined, and this is the composition that satisfies the target property. Wider the range of S or C_s , more capable it will be to deal with the changes in the required property in the future, and such breadth is preferable. In practice, the cost and tolerance are also considered within this composition to determine the final composition.

4 Weak conditioned combinatorial linear programming system

The aforementioned mathematical process is too complicated for

a material engineer to conduct as a daily routine, and the volume of computation becomes extremely large when n increases because it is necessary to calculate the solution space for each two combinations. It is nearly impossible to calculate the solution space by Excel or by hand. Therefore, we developed the solution search software M-Designer that can be used practically at the site of material development.^{[10]-[12]} The M-Designer is a weak conditioned combinatorial linear programming software with interface shown in Fig. 6. Using the M-Designer, it is possible to compute whether there is a composition (solution set) that satisfies the target value for each combinatorial index. Specifically, the maximum and minimum values of the property/composition matrix, range of target property, number of compositions, and content that were obtained experimentally or theoretically are entered. The contents of the material are automatically changed at appropriate intervals, decision is made whether it satisfies the target, and the compositions that satisfy the target value are provided.

The composition candidates must simply satisfy the target value, and it is not necessary to seek the optimal value. If the range is wide, the possibilities of various usages increase. Since it is a simple system without the implementation of the algorithm to calculate the optimal value, we found that it could be used widely for nonlinear programming including complex physical property values as explained later, as well as for compositional design including the time-series data.

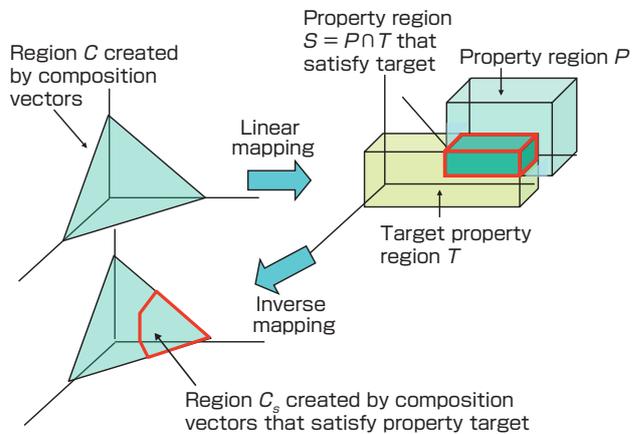


Fig. 5 Outline of the weak conditioned linear programming

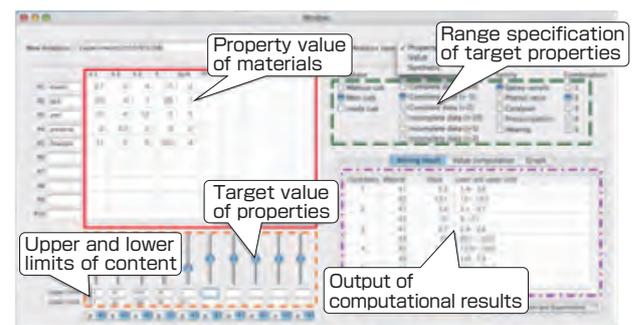


Fig. 6 System interface

Figure 7 shows an example of a simulation. The comparison of the actual results had been reported,^[13] but the property can be roughly predicted, although it may lack precision. To increase the precision, the relational equation between the composition content and property should be preferably expressed by nonlinear simultaneous equation. However, in the case where several combinations of materials must be investigated, extremely large amount of experimental data becomes necessary to calculate the function of n variables for each combination. In practice, according to the procedure shown in Fig. 8, the candidate materials are narrowed down by linear approximation, and the nonlinear approximation is conducted only when precision is required.

This system was used for investigating the adhesive agent composition where several materials were combined. As shown in Fig. 9, it was found that wide ranges of functional extensions and applications were possible such as extension to nonlinear programming,^[14] calculation of material supply risk,^[13] and database building.^[15]

With adhesive tapes and adhesive agents, if there is gap θ in phase between the stimulus and response of the material, for the strain expressed by the sinusoidal function $x = Ae^{-i\omega t}$, the

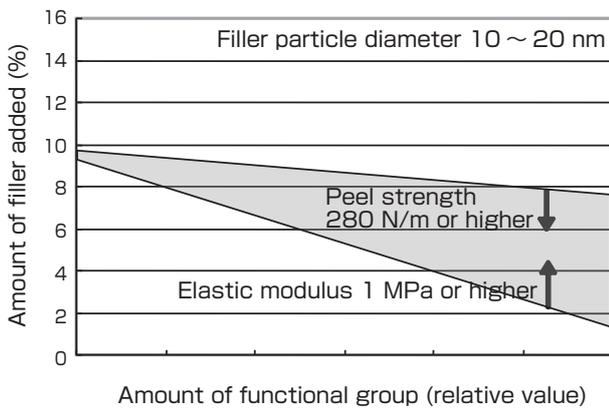


Fig. 7 Example of simulation

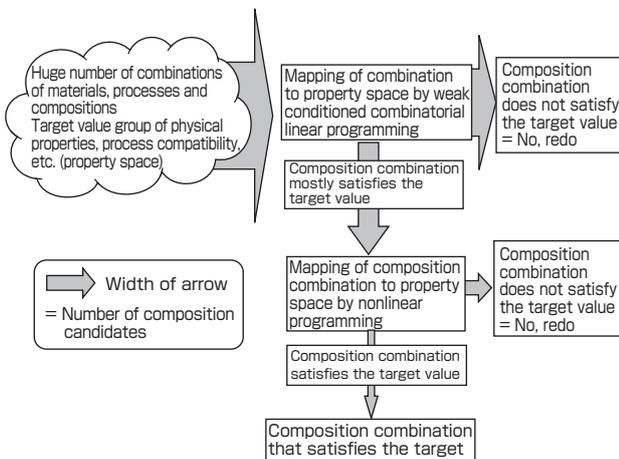


Fig. 8 Distinction of linear and nonlinear approximations

property value (elastic modulus) defined between the two becomes $y/x = (B/A) \times e^{i\theta}$, when the stress is $y = Be^{-i(\omega t - \theta)}$. Then, the property value becomes a complex number and its control is extremely important. In this case, the relationship between the composition and property becomes a complex nonlinear function. Such complex property value can also be processed by the aforementioned system by handling the complex number as the vector of the Gaussian plane.^{[10][16]}

One of the points that became apparent in the Great East Japan Earthquake in 2011 was the risk of material supply cutoff. In the highly efficient and aggregated semiconductor and automobile industries, the cutoff of a single material or part can have major effects. Using this system, it is possible not only to calculate the candidate compositions, but also calculate the supply risks, considering whether the multiple candidates use a common material. Although there is the issue that the material supply risk cannot be accurately grasped, if it is assumed that there is a certain risk for each material such as 0.01 %, the index that expresses the supply robustness of the composite product can be calculated. It is then possible to consider the risk countermeasures in the design phase.

Such risk calculation was not possible in the conventional linear programming, and it became only possible with the weak conditioned combinatorial linear programming and the construction of its computation system. This result demonstrated the effectiveness of this method.

The database that can organize and store the data for property/composition matrix is useful in utilizing the past failure data in the new development. If only the past experimental data were stored, it would not be very useful

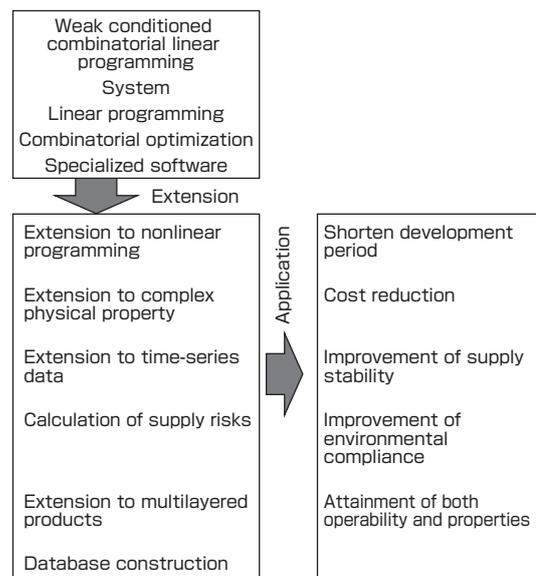


Fig. 9 Extension of weak conditioned linear programming and its application range

since the correlation between the property and composition is unknown, but the property/composition matrix data can be stored, entered into the M-Designer, and calculation can be conducted for reuse. For example, although something was a failure in the past, with major changes in structure and target values, the composition could satisfy the target value with some corrections. Such database can prevent repeating the same mistakes because one did not know the past results.

5 Application of the weak conditioned combinatorial linear programming

Since this method is particularly useful for short-term development, it is not only useful for the development of semiconductor materials, but can also be applied to various fields such as foods and environment in the future.

In this paper, focus was placed on the optimization of composition, but the design method is not limited to composition. Instead of the composition content, various manufacturing conditions other than substances, such as temperature, humidity, or speed, can be considered.

The optimization method of this paper can be applied to various fields in the future. For example, there is an endless list of fields where combinatorial optimization is important, such as cooking and blending perfume, alcoholic beverages, or spices, as well as drugs such as herbal preparation and synthesized drugs. Particularly in the food industry, the design of taste by the combination of flavors or ingredients is an extremely important theme. In practice, the cook determines the taste according to his/her expert perception and experience, such as changing the flavoring to utilize the seasonal ingredients, and mathematical design has almost never been done. With the development of the taste sensor and the quantitative evaluation of taste, we predict that the mathematical design will greatly advance in this field in the future.

6 Conclusion

In this paper, we described the material design method constructed to increase the design efficiency of the semiconductor-implemented materials. By utilizing this method, it became possible to conduct on-demand product development with adjustability, or the ability to match the target value by flexibly changing the material properties.

Because breakthrough is not the objective, our method may be criticized as merely being a partial optimization that is a diversion from larger work. Yet, breakthrough can be sought by someone else. If there is no compromise, short-term development will not be possible.

The site of short-term material development was

considered alien to serendipity. However, when we conduct experiments after predicting the properties from the material combination using this method and clarifying the baseline, we unexpectedly became capable of detecting the deviations from the prediction. While most cases are errors in prediction, in rare instances, some were synergetic effects of unforeseen, new structures. We are now able to detect the hints of serendipity.

We believe that with more feedback, the new material design method and the system proposed in this paper will become an essential tool for the timely development of necessary materials. The ultimate goal is to create an on-demand material development system that is fused with craftsmanship.

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Japan, and others. In this paper, contributed to the foundation of weak conditioned combinatorial linear programming and the development of its software.

Discussions with Reviewers

1 Overall

Comment (Hideto Taya, Public Relation Department, AIST)

This paper discusses the effort for building a tool to support material design for developing materials with target performance within a limited time, by combining the existing materials at the manufacturing sites of state-of-the-art products. It is very interesting, and the content matches the objectives of *Synthesiology*.

2 Style of material development

Question (Hideto Taya)

At the manufacturing sites of the state-of-the-art semiconductor materials, in the constraint of an extremely short time limit, the on-demand material development method has been devised "to develop materials that achieve the target value in a practical and certain manner rather than by simple *bricolage*."

Is the on-demand material development method an add-things-together development that is merely practical but not creative, or is it a "method that combines the merits of *bricolage* and engineering (*aufheben* in Fig. 1)"? How will the relationship between the two develop in the future?

Answer (Teiichi Inada)

The style of material development in this paper is, first, an add-things-together development that is merely practical but not really creative. However, when this is taken to the extreme, unexpectedly, we were able to discern unforeseen synergetic effects. This is explained by the diagram for *aufheben* of practicality and creativity, as shown in Fig. 1.

For example, when properties were predicted from the material combinations using our method and experiments were done after clarifying the baseline, we realized we could detect deviations from the predictions. That meant, by predicting the experimental results without synergetic effect, we became capable of detecting small synergies. There were examples in which after discovering small synergetic effects, we succeeded in increasing the synergies and created a differentiating technology. Although the material development style in this paper seem to be cut-and-dry development conducted under time constraints, it can also play the role of a microscope to discover unforeseen effects.

3 Structure of the paper

Question (Hideto Taya)

For chapter 3 "Basics of the weak conditioned combinatorial linear programming," how about setting up new subchapters, and add more detailed explanations for simulation results, different uses of linear and nonlinear, and expandability?

Answer (Teiichi Inada)

I revised the chapters and increased the explanations. The content is included in the paper, and here, I shall explain the main points only.

1) Expandability to complex numbers

With adhesive tapes and adhesive agents, the property values often become complex numbers. It was clarified that such complex physical properties could be processed using the aforementioned system by handling the complex numbers as vectors of the Gaussian plane.

2) Calculation of the supply risk

It is important that the materials can be supplied stably, and not just satisfy the target properties. In cases where there were risks of material supply cutoff, we considered whether the composition candidates used common materials, and calculated the supply risk. As a result, we were able to estimate the supply risk along with the material that satisfied the targets. Such risk calculations could not be done with conventional linear programming, and it became possible for the first time with the construction of the weak conditioned combinatorial linear programming and the computation system.

3) Database construction

By storing the past experimental results and data in the form of property/composition matrix, such data can be utilized effectively by entering them in the M-Designer and then reusing them for calculation. For example, although they were failures in the past, there were cases where because there were major changes in the structure and target values, the target value could be satisfied with only slight revisions. The development efficiency can be improved by using the past results.

4 What are the problems of this development method?

Question (Hideto Taya)

What were the evaluations and comments when applying the on-demand material development method at the actual production sites? Were there any points that you particularly considered in the implementation?

Answer (Teiichi Inada)

When we explained this method at the company, a senior engineer remarked, "If you write out all that goes on in the head of a veteran engineer, you will have something like this." Certainly, a veteran engineer, who is thoroughly versed in the good and bad of a material, designs things so the bad part stays in the background and the good part floats to the foreground. When

we pursued material development in a rational manner, we ended up at craftsmanship. This is not surprising, and it shows how craftsmanship is systematic and excellent. In the future, I think we can construct a system that goes one step further, by sublimating and integrating the rationalization and craftsmanship.

5 Shortening of the development time by using this method

Question (Hideto Taya)

You say, "This method is particularly effective for short-term development." Based on your experience, how much was the development time shortened?

Answer (Teiichi Inada)

Since target values and conditions differ each time in the actual development, it is difficult to accurately compare the cases where this method was used or not used. However, based on experience, there are the following merits:

1) There are myriad composition candidates, but by using this method, first, the compositions that are totally off-target could be eliminated, and the choices can be narrowed down to highly potential candidates only. Since this method is a linear approximation, there is certainly a deviation from the experimental data, but it is sufficient for narrowing down the candidate materials, and the development time can be shortened greatly.

2) The product improvements are modified based on the past results. For example, when developing a new product by revising a product made by our predecessors 10 years ago, there are cases where we have good performance although we do not know why some materials were included. In such cases, we do not know why we get a certain function or why this certain chemical is needed, and time must be taken even for slight modifications. In such cases, the analysis system and database are extremely useful to shorten the development time.